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STUDY OF OPTICAL SOUND GENERATION
AND AMPLIFICATION

Annual Report

PARGUM 86-04

1 November 1985 - 31 October 1986

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bу

Henry E. Bass and F. Douglas Shields Physical Acoustics Research Center The University of Mississippi University, Mississippi 38677



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20. ABSTRACT (Continue on reverse side if necessary and identify by block number)

This project involves three separate tasks with the following titles: Task I \sim Generation of low frequency sound from optical pulses; Task II - Propagation of sound through a gas with an overpopulation of vibrationally excited states; and Task III - Optoacoustic studies in liquids. The three tasks, funded by the physics division of ONR, represent a three-pronged study of the generation of sound by the absorption of light and amplification of a propagating sound wave.

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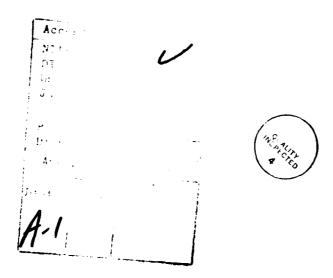
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20. ABSTRACT CONTINUED

Generation of low frequency sound from a series of optical pulses involves optimizing the density of pulses to obtain maximum energy at low frequencies. The present effort is devoted to numerical simulation. With present modulation schemes, as much as 50% of the energy appears in the desired fundamental, but predicted harmonic distortion is unacceptable.

Amplification of sound propagating through a gas with an overpopulation of vibrationally excited states has now been observed in $\rm N_2/H_2$, $\rm N_2/He$, and $\rm N_2/CH_4$ mixtures. The gas is excited with an electrical discharge. Present efforts are devoted to achieving better agreement between theory and experiment.

Optoacoustic pulses have been observed in several alcohols and water. The optoacoustic amplitude has been observed as a function of laser energy, distance from excitation zone to detection region, and optical absorption coefficient.



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Abstract

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The optoacoustic amplitude has been observed as a function of laser energy,

distance from excitation zone to detection region, and optical absorption

coefficient.

Generation of Low Frequency Sound from Optical Pulses

Most optical sources efficiently produce short pulses of light with constant pulse height and width. It is generally believed (but not yet proven) that the optical/acoustic conversion efficiency upon absorption increases with decreasing pulse length. For individual pulses, very short pulse lengths suggest high frequency sound which is attenuated rapidly by a fluid. we are investigating the use of a train of high frequency pulses with a pulse density proportional to the desired acoustics amplitude and rely on known attenuation mechanisms to absorb the high frequency components of the optically generated acoustic pulses. We refer to this technique as pulse density modulation (PDM).

A preliminary computational study of this modulation technique was completed under the present contract and the results reported in an Annual Summary Report, 15 January 1983. In summary, that study illustrated that a pulse repetition frequency in excess of 6400 times the acoustic frequency is necessary to achieve an S/N ratio in excess of 20 dB (considered the minimum acceptable). Conversion efficiencies near 50% are possible with this PRF. For the purposes of these computations the pulse width was one unit (1/6400 of the acoustic period) wide.

The previous modulation scheme placed the acoustic pulse in time according to the amplitude of $1 + \sin \omega t$ where ω is the desired acoustic frequency. More recent work has been devoted to computer simulations to determine if this placement is optimum. This numerical procedure takes Np pulses and places them in N positions comprising a single cycle of the acoustic wave. Though this process is straightforward, it requires extensive computer time (about 2 hours of mainframe time per Np, N combination). There are not yet sufficient results to draw conclusions.

The Propagation of Sound Through a Gas with an Overpopulation of Vibrationally Excited States

Several years ago, the SACER effect had been predicted^{1,2} and there were experiments indicating it existed in gases reacting chemically^{3,4}. However, there had been no observations of the effect in gases with an overpopulation of vibrationally excited states. We undertook the ambitious task of not only observing this phenomenon, but accurately measuring the magnitude of the effect and comparing it to calculated values.

The first step was to theoretically analyze a case that could be simulated in the laboratory. Previous efforts had been made to observe the effect while a mechanism was taking place pumping molecules into the excited state. In order to simplify the analysis, we looked for a situation where a metastable vibrational state persisted long enough after the pumping mechanism was removed to allow the passage of a sound wave through the gas. Theoretical calculations of the magnitude of the expected sound amplifications for this situation in some specific gases were published in 1984.

These calculations showed that some very stringent conditions must be met if the effect was to be experimentally observed. For the magnitude of the gain to be appreciable, the v-t relaxation time could only be about one order of magnitude longer than the sound period. A number of experimental configurations have been tried to meet these conditions. Suffice it to say here that the effect has now been observed and measured in N_2/H_2 , N_2/He and N_2/CH_A mixtures.

Measurements of the absolute magnitude of the amplification as yet do not agree with calculated values, but the general dependence of the gain upon the vibrational relaxation time and vibrational temperature is as expected from theory.

Another discovery has been made in these experiments that was not expected. We have been able to measure the variation of the sound velocity in the gas as the vibrational energy decays into translation and from these measurements determine the variation of the translational temperature during the relaxation process. This in turn, allows a determination of the vibrational temperature as a function of time and, thus, the vibrational relaxation time and its temperature dependence. In the diatomic gases currently under study (N_2 and CO) these numbers are very difficult to measure otherwise and constitute a valuable addition to the literature. The results of the N_2/H_2 measurements are reported in a paper recently submitted to JASA and the measurements in N_2/He , N_2/CH_4 mixtures will be reported at the December meeting of the Acoustical Society or America. In the case of N_2/CH_4 mixtures, the effect of spark-induced-reaction contaminants on the relaxation time in the gas mixture is evident.

From the results obtained to date, it is concluded that the SACER phenomena needs to be considered in many non-equilibrium situations. It is likely involved in producing instabilities in excited gases in a variety of circumstances.

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Optoacoustic Studies of Liquids

Optoacoustic generation of acoustic pulses in a fluid has been widely studied from a phenomenological point of view. It is found 1,2,3, for example, that for low optical energies, the acoustic pulse generated is a result of thermal expansion. For larger energies, vaporization and electrostriction can be important. It is the purpose of this study to explain these phenomena in terms of the microscopic properties of the field.

In order to view results on the microscopic level, we chose the model of the fluid as being composed of molecular groups, clusters, with some average, though dynamically changing number of molecules per cluster. A liquid in a given volume, V, has Nc clusters. A pulse of laser light incident upon this volume has Np photons where Np depends upon pulse energy and wavelength. We further assume that for every photon absorbed, a molecule (which can be more than one or a constant fraction) is disassociated from its cluster. This localized density variation is the origin of the acoustic pressure wave.

The amplitude of the acoustic pulse is determined by the number of molecules stripped from their associated cluster. This is a microscopic description of thermal expansion. So long as

$$\frac{Nc}{Np} > 1$$
,

that is, the number of clusters exceeds the number of photons, the amplitude of the acoustic pulse will vary linearly with input energy.

The experimental investigation of this phenomena was patterned after Tam⁴. The acoustic pulse generated by a high peak power pulsed excitation laser is observed by the deflection of a CW probe laser beam. The excitation and probe beams are aligned such that they are almost parallel as they

pass through the test cell containing the liquid of interest. The probe beam impinges upon a fast photodiode such that the detector intersects only a wing of the Gaussian-shaped probe beam. A deflection of the probe beam across the photodiode will then generate a variation in diode output which is displayed on an oscilloscope. The recorded signal is actually proportional to the pressure gradient across the probe beam as a function of time. The stored signal is transferred to a PDP 11/23 computer interfaced to a 7854 Tectronix scope of simply photographed directly from the CRT.

The excitation laser is a PRA nitrogen laser emitting 800 ps pulses at a wavelength of 337 nm. The temporal pulse shape is approximately Gaussian. The excitation beam has a rectangular shape which, when focused, gives a size of about 50 μ m x 15 μ m. The maximum intensity at the quartz test cell was 2.27 x 10^{14} W/m². The probe beam (CW He-Ne) is spatially filtered and focused to a diameter of 10 μ m at the test cell.

The test liquid used in most of the work was propanol doped with red dye to increase the absorption coefficient α . By adding drops of dye to the propanol and recording the energy transmission for each additional drop, α can be calculated for the liquid as a function of the number of drops. Once α is determined, the penetration depth of the laser pulse can be controlled. We typically added dye until the penetration depth was $\tilde{}.87$ mm. The excitation volume could then be determined by the area of the beam and the penetration depth. The power per unit volume for this penetration depth was 2.6×10^{17} W/m 3 .

The first experiment conducted was designed to test linearity. by placing a fixed number of glass slides in the path of the excitation beam, the power delivered to the test volume was varied in steps from 2.6 x 10^{17} W/m 3 to 2.13 x 10^{16} W/m 3 . The results are shown in Figure 1. It is obvious that the optoacoustic signal amplitude varies linearly with energy.

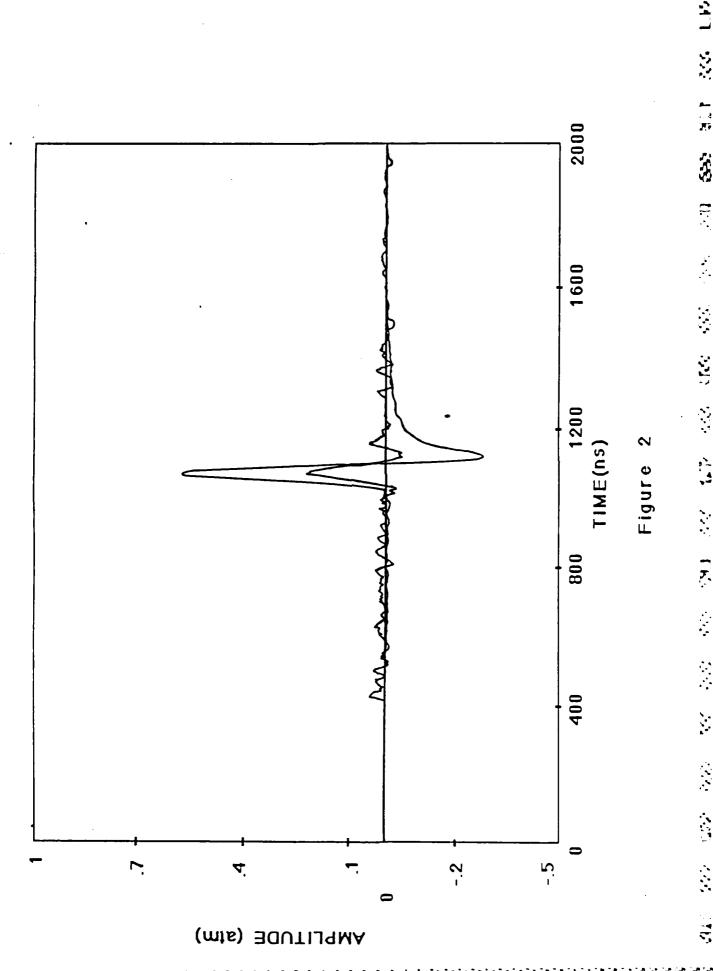
The second measurements involved calibration of the probe beam deflection by comparing the amplitude of signal deflection to the output of a calibrated hydrophone. The hydrophone used had a bandwidth from 1 to 10 MHz. The amplitude and time evolution of the hydrophone signal is compared to a theoretical calculation in Figure 2. Although agreement is not exact due to the bandwidth of the transducer, we consider this reasonable validation of the theoretical treatment given the frequency response of the hydrophone.

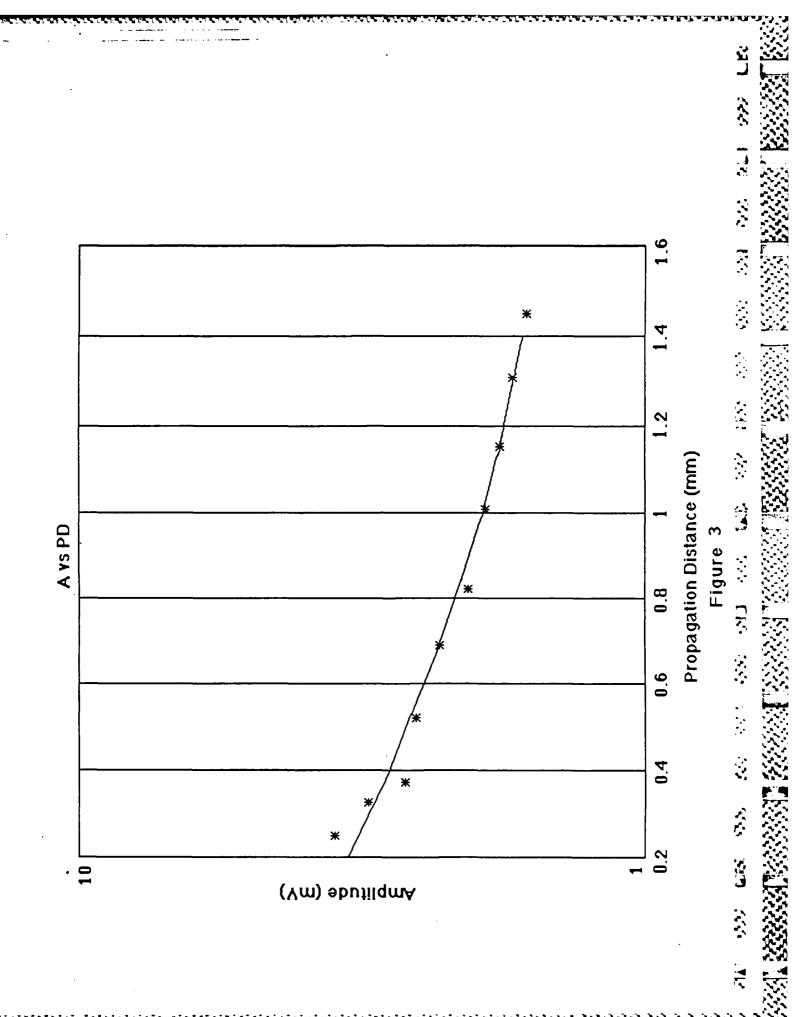
The final result to report is the variation of the optoacoustic signal amplitude with propagation distance. A typical result is shown in Figure 3. It can be seen that for this range of propagation distances, the signal falls off as $1/\sqrt{r}$, suggesting cylindrical spreading. We would expect to observe 1/r decrease at propagation distances greater than a few mm.

The results to date suggest that the optoacoustic signal is a result of thermal expansion for intensities where the number of photons is about 1/16 the number of clusters or less. This is not surprising since for the pulse length used, one would expect significant recombination during the duration of the laser pulse. Future work will be devoted to shorter pulses with subsequently greater peak powers in order to define conditions for transition to nonlinear response.

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STUDY OF OPTICAL SOUND GENERATION AND AMPLIFICATION

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GRADUATE STUDENTS SUPPORTED UNDER CONTRACT FOR YEAR ENDING 30 SEPTEMBER 1986

Yijian Cao Stanley Cheyne Charles Thompson Wei Xie

POSTDOCTORALS SUPPORTED UNDER CONTRACT FOR YEAR ENDING 30 SEPTEMBER 1986

None

PAPERS PUBLISHED IN REFEREED JOURNALS

"Spectrophone Measurements in Sulfur Hexafluoride," M. H. Ali, H. E. Bass, and H. X. Yan, IEEE Trans. on Ultrasonics, Ferroelectrics, and Frequency Control UFFC-33, 615-621 (1986).

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PAPERS SUBMITTED TO REFEREED JOURNALS (Not yet published)

F. Douglas Shields, "Propagation of Sound in Vibrationally Excited $\rm ^{N}_{2}/\rm ^{H}_{2}$ Mixtures," J. Acoust. Soc. Am., November 1986.

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BOOKS (AND SECTIONS THEREOF) PUBLISHED

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STUDY OF OPTICAL SOUND GENERATION AND AMPLIFICATION

November 1, 1985 - October 31, 1986

Status Report

Graduate Students and Postdoctoral Personnel Associated with Contract

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Other Government Sponsored Research -- Principal Investigator(s) Associated

Henry E. Bass

Construction Engineer- Graduate Student Support Aug 86-Aug 87 ing Research Lab

Army Research Office Long Range Sound Propaga- Dec 83-Dec 86 tion in the Atmosphere

Waterways Experiment Remote Acoustic Detection Mar 86-Sep 86

Waterways Experiment Remote Acoustic Station of Buried Mines

Current Status of Contract Funds

Contract ends December 31, 1986 with no remaining funds anticipated.

Permanent Equipment Purchased

Nitrogen Pulsed Laser System
High Speed Sampling Oscilloscope
Optical Table
Picosecond Photodetector
Nitromite Dye Laser
Analog Filter
Wavetek Model 442
Spatial Filter Assembly

DESCRIPTION OF A STATE OF A STATE